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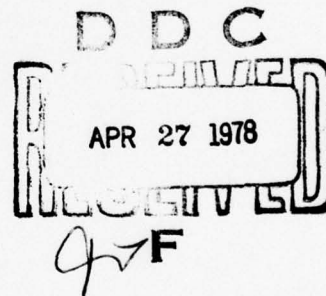
ELASTIC VERSUS MAGNETOELASTIC ANISOTROPY IN RARE EARTH- IRON ALLOYS

BY J. R. CULLEN
S. RINALDI
G. V. BLESSING

RESEARCH AND TECHNOLOGY DEPARTMENT

1 NOVEMBER 1977

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SUMMARY

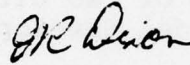
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Finally a transmission experiment in a single crystal ($\text{Tb}_{.3}\text{Dy}_{.7}\text{Fe}_2$) is described. Resonant-like transmission is shown to take place when an applied magnetic field is so oriented as to produce almost equal velocities for the C_{44} and $C_{11}-C_{12}$ modes.

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PREFACE

The work reported here represents a portion of the research carried out by the Magnetism and Materials Group, Solid State Branch, Research and Technology Department, to determine the elastic properties of highly magnetostrictive rare earth-iron alloys. The work was supported by the Office of Naval Research (Task No. RR011-08-01). The report summarizes results on single crystal, polycrystalline and amorphous alloys.



J. R. DIXON
By direction

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INTRODUCTION

Magnetically ordered solids generally undergo a change in length relative to their length in the demagnetized condition. This relative length change, or magnetostriction is an intrinsic property of the magnet and varies in size from nearly one percent in ferromagnetic Rare Earth metals at low temperatures to a few parts in a million in the ferromagnetic transition elements Co, Fe and Ni. [1] Magnetostriction results from the coupling of the magnetic moments to the lattice distortions. This same interaction is capable of modifying the elastic properties of magnetostrictive materials. Besides the changes in modulus brought on by domain-wall movements such as the ΔE effect [2], there are often measurable changes in the elastic properties in magnetically saturated materials. The latter effects are most clearly observable in single-crystal specimens. Changes in elastic constants are most pronounced in highly magnetostrictive magnets. In Ni for example, changes in elastic constants of the order of 10^{-3} have been observed [3]; in rare earth metals these changes can be several percent [4].

1. Clark, A. E., in Handbook of the Physics and Chemistry of the Rare Earths, K. Gschneidner and H. Eyring, eds. North Holland (Amsterdam), to be published.
2. Bozorth, R. M., Ferromagnetism, Van Nostrand 1951.
3. Alers, G. A., Neighbours, J. R., and Sato, H., J. Phys, Chem. Solids 9, 21 (1958).
4. Moran, T. J., and Luthi, B., J. Phys. Chem. Solids 31, 1735 (1970).

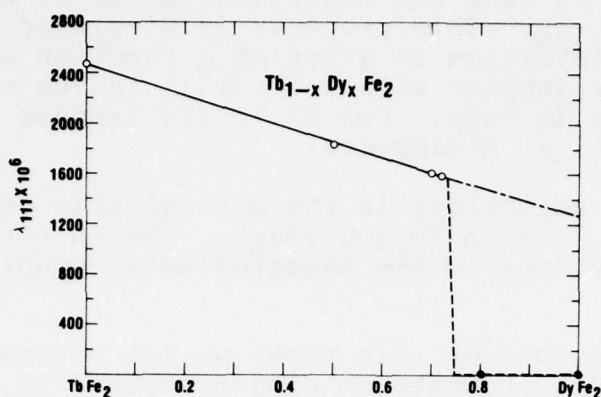


Fig. 1 Magnetostriction constant λ_{111} determined by the x-ray method.

For ferromagnets, an estimate for the change in elastic constant can be made from the formula

$$\delta C/C = -\lambda^2 C/K. \quad (1)$$

Here C is a modulus, λ the magnetostriction and K the magnetic anisotropy. For Ni, $C \approx 10^{12}$ erg/cm³, $K \approx 10^5$ erg/cm³, $\lambda \approx 10^{-5}$ and Equation (1) gives $\delta C/C \approx 10^{-3}$. That the changes in modulus are so dependent on magnetic anisotropy simply reflects the fact that the slowing of a sound wave is accomplished by a rotation of the moment.

Recently, attempts have been made to produce materials which would have the high magnetostriction characteristic of the rare earths but have this property available at room temperature. Success was achieved [1] with the cubic rare earth-iron compounds RFe_2 , where R stands for a rare earth element. In $TbFe_2$ for example, $\lambda_{111} \approx 2.6 \times 10^{-3}$ at room temperature. Since this is two orders of magnitude larger than the magnetostriction in Ni, we would expect modulus changes of several percent in these compounds were it not for their characteristically large magnetic anisotropy. The latter difficulty can be overcome with little expense in magnetostriction, however, by forming pseudobinary compounds $R_{1-x}R'_xFe_2$ with a suitable choice of x . In $Tb_{0.3}Dy_{0.7}Fe_2$, for instance K is less than 10^6 erg/cm³ while λ is still large (1.6×10^{-3}). The reason this tailoring of λ and K works is that these two quantities depend almost exclusively on the rare earth constituent. Using these two values, the formula gives $\delta C/C \approx -1$ at room temperature. This was more than ample motivation for an ultrasonic investigation of the elastic constants of this type of pseudobinary compound.

One other characteristic property of these compounds is of special interest here. That is that the magnetostriction is highly anisotropic: $\lambda_{111} \gg \lambda_{100}$. This property is displayed in Figure 1 in which the x-ray distortion is given as a function of x in $\text{Tb}_{1-x}\text{Dy}_x\text{Fe}_2$. [5] For samples with $x < .7$, [111] is the easy axis and the distortion (λ_{111}) is huge. For $x > .7$, the samples have [100] easy and the distortion (λ_{100}) disappears.

We expect little anisotropy in the pure elastic constants ($a \equiv 2 C_{44}/C_{11}-C_{12} \approx 1.25$ in $\text{Tb}_{.3}\text{Dy}_{.7}\text{Fe}_2$). The anisotropy in λ then translates into anisotropy in the magnetoelastic coupling: $b_2 \gg b_1$.

In succeeding sections of this paper we (a) review our recent ultrasonic velocity measurements on single-crystal $\text{Tb}_{.3}\text{Dy}_{.7}\text{Fe}_2$ from which the elastic constants and b_2 were determined; (b) discuss recent results of velocity measurements in highly magnetostrictive polycrystalline and amorphous rare earth-iron alloys. Finally, we describe a measurement on single-crystal $\text{Tb}_{.3}\text{Dy}_{.7}\text{Fe}_2$ of a resonant coupling of the C_{44} mode to the $C_{11}-C_{12}$ mode which is a direct consequence of the anisotropy in magnetostriction and the fact that $\delta C/C > a-1$.

SINGLE CRYSTAL RESULTS: C_{44} and $C_{11}-C_{12}$

The velocity of transverse sound waves was measured in a single crystal of $\text{Tb}_{.3}\text{Dy}_{.7}\text{Fe}_2$. [6] The direction of propagation was [110]. All our measurements were done at room temperature, using frequencies near 10 MHz. Figure 2 shows the results of applying a magnetic field of up to 25 kOe in various directions for both [001] (C_{44}) and [110] ($C_{11}-C_{12}$) polarizations of the sound. Note that the velocity of [001] waves has been reduced by thirty percent upon rotating the field in the (110) plane from perpendicular to the polarization to parallel to it. This corresponds to more than a 50 percent reduction in C_{44} as we anticipated from the estimate given above. The velocity of the $C_{11}-C_{12}$ waves was increased by a percent or so as the magnetic field was rotated from perpendicular to the polarization to parallel to it. From the form of the magnetoelastic energy in terms of the magnetization components M_i and the strains ϵ_{ij} for cubic systems, ie. we see that b_2 couples components of the moment to C_{44} -type

$$E_{me} = \frac{b_1}{M_s^2} \sum_i (M_i^2 - 1/3) \epsilon_{ii} + \frac{b_2}{M_s^2} \sum_{i \neq j} M_i M_j \epsilon_{ij} \quad (2)$$

5. Clark, A., Cullen, J., McMasters, O., and Callen, E., A.I.P. Conf. Proc. No. 29, (A.I.P. New York) p. 129.
6. Rinaldi, S., Cullen, J. R., and Blessing, G. V., Phys. Lett. 61A, 465 (1977).

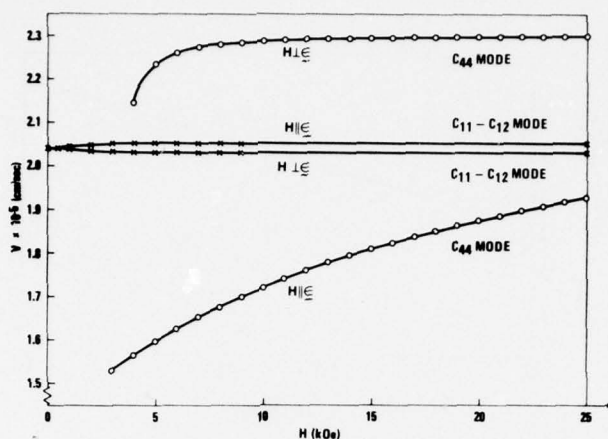


Fig. 2 Sound velocity vs. magnetic field: polarizations along [001] (circles) and along [110] (crosses). For each polarization, data for two orientations of field are shown.

(ϵ_{ij} $i \neq j$) strains while b_1 is appropriate for $C_{11}-C_{12}$ ($\epsilon_{ii}-\epsilon_{jj}$) strains. Thus the large changes in C_{44} and relatively small ones in $C_{11}-C_{12}$ confirm the results of earlier work that $b_2 \gg b_1$.

From Equation (2) and the quasi-static approximation [7], which is good whenever the sonic frequencies are much less than the ferromagnetic resonance frequency, it is possible to calculate the change in the elastic energy in terms of the susceptibility tensor and the components of the moment and magnetic field. For $H \parallel [001]$ the change in C_{44} is predicted to be (H^0 is the total internal field,)

$$C_{44} = C_{44}^0 - b_2^2 / (H + H^0) M_S \quad (3)$$

i.e. the sum of the anisotropy field and $4\pi M_S$ in the case of conducting materials like these.

Here C_{44}^0 is the pure elastic constant. The change in C_{44} for $H \parallel [110]$ is zero; thus $C_{44}(H \parallel [110]) = C_{44}^0$ and by plotting $(C_{44} - C_{44}^0)^{-1}$ vs. H , $|b_2|$ was determined from the slope of the resulting straight-line portion (see Figure 3) to be 2.3×10^9 erg/cm³ in excellent agreement with earlier measurements. [1]

7. See for example, Le Craw, R. C., and Comstock, R. L., in Physical Acoustics ed. W. Mason Vol III B page 127 (Academic Press 1965).

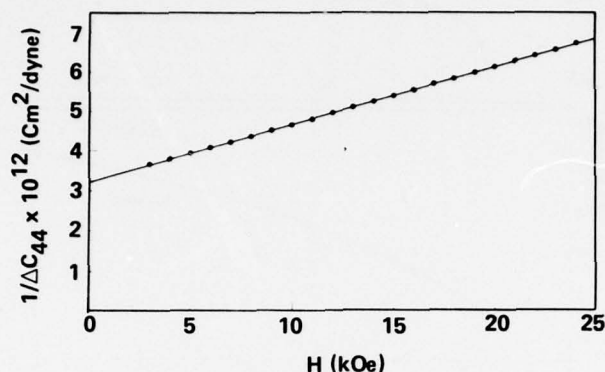


Fig. 3 The reciprocal of the difference in C_{44} , $(C_{44}^0 - C_{44})^{-1}$ plotted vs. magnetic field. b_2 is determined from the slope.

Further verification of the magnetostrictive origin was obtained from a plot of $(C_{\max} - C)/(C_{\max} - C_{\min})$ versus the angle of the magnetic field with respect to the crystalline axes (Figure 4). The high-field limit of the theory [8] based on (2) is the solid line in this figure. We see that the over-all agreement is quite good, better at 10 kOe than at 6 kOe. This is understandable considering that the theoretical curve is a high-field limit. Returning to the $(C_{44}^0 - C_{44})^{-1}$ vs. H plot, from the intercept and (3) we obtained the magnetic anisotropy. Using 800 Oe for M_S we found $K = 4 \times 10^6 \text{ erg/cm}^3$, about eight times greater than the value obtained from more conventional methods. [9]

We mentioned that the velocity of the C_{11} - C_{12} type mode actually increased slightly when according to (2), it should have decreased (see Figure 5). This is evidence for a non-linear term in the magnetoelastic coupling. Since the shift in C_{11} - C_{12} is independent of applied field, the non-linear coupling [10] is probably the "morphic" type, [11]

$$E'_{me} = F M_i M_j \epsilon_{kl} \epsilon_{mn} \quad (4)$$

8. Simon, G., *Naturf, Z.*, 13A 84 (1958).
9. Williams, C., and Koon, N., Conference on the Rare Earths and Actinides, *J. Phys.* (to be published).
10. Bonsall, L., and Melcher, R., *Phys. Rev.* 14, 1128 (1976); Dohm, V., and Fulde, P., *Z. Phys.* B21, 368 (1975).
11. Mason, W., *Phys. Rev.* 82, 715 (1951).

where F is a tensor of rank six. We will return to this question of non-linearity in the final section.

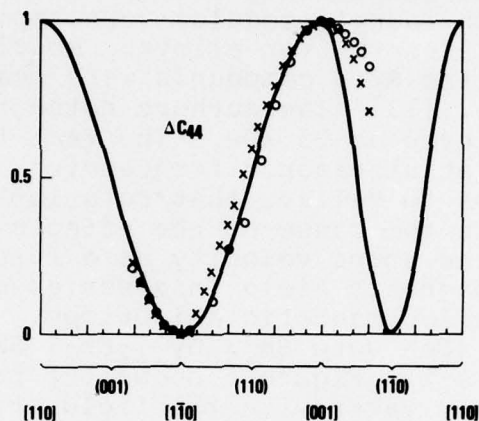


Fig. 4 Angular variation of the relative change: $(C_{\max}-C)/(C_{\max}-C_{\min})$ in elastic constant (C_{44}) in the (001), (110) and ($\bar{1}\bar{1}0$) planes. Circles: 6 kOe data. Crosses: 10 kOe. The full lines are a result of linear magnetoelastic theory.

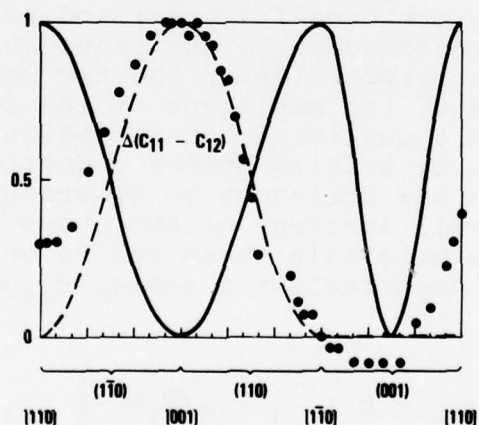


Fig. 5 Angular variation of the relative change: $(C_{\max}-C)/(C_{\max}-C_{\min})$ in elastic constant ($C_{11} - C_{12}$) in the ($\bar{1}\bar{1}0$), ($\bar{1}10$) and (001) planes. All data taken at 10 kOe. Dashed lines correspond to the morphic effect; full lines correspond to linear magnetoelastic theory.

POLYCRYSTALLINE AND AMORPHOUS MATERIALS: ELASTIC ISOTROPY

Changes in moduli in magnetostrictive polycrystalline materials are very often observed and usually attributed [2] to the movement of domains. Changes in Young's modulus were reported earlier at low fields [12] in rare earth-iron alloys. Modulus changes in a series of polycrystalline RFe₂ compounds were measured at 10 MHz in fields up to 25 kOe. [13] The authors noted that in TbFe₂ saturation was not achieved even at 25 kOe. The fact that such large changes were observed at ultrasonic frequencies, too high for domain walls to follow, led us to believe that rotation of the magnetization by the strain field was the cause of the effects. We therefore made [14] a study of the sound velocity as a function of the magnitude and orientation of a magnetic field in a series of highly magnetostrictive polycrystals with low magnetic anisotropy. Included in the study, besides Tb₃Dy₇Fe₂ were Sm₈₈Dy₁₂Fe₂, Sm₇Ho₃Fe₂ and amorphous TbFe₂ (a-TbFe₂). Figure 6 shows the pattern of echoes we observed from a-TbFe₂ taken with the field at various angles θ with respect to the direction of polarization of the transducer. For $\theta = 0^\circ$ or 90° we observed a single train of echoes, though those at 0° were delayed relative to those at 90° . At intermediate θ , however, we observed a superposition of the 0° and 90° patterns indicating that two shear waves are propagating simultaneously, with different velocity, in these configurations. It is clear from Figure 6 that the height of the shorter-time pulse (amplitude of the faster wave) decreases as θ decreases from 90° , while the delayed-pulse height (slow-wave amplitude) increases. We checked our interpretation of the double echo pattern by making a through transmission experiment with the transmitter and receiver shear polarizations mutually orthogonal ($\epsilon_T \perp \epsilon_R$) and rotating the magnetic field in the plane perpendicular to the propagation. Using a large pulse width to allow superposition of the two waves, the expected $\sin \theta \cos \theta$ dependence of the amplitude of the detected signal was verified. We obtained a qualitative description of the two waves simultaneously present by writing down a coupling between strain and magnetization in a way analogous to Equation (2), but taking into account the over-all isotropy of amorphous and polycrystalline matter. For isotropic materials there can be only one coupling constant b , since the magnetoelastic energy E_{me} must be rotationally invariant. Thus

$$\bar{E}_{me} = b \sum_{i,j} \epsilon_{ij} (\bar{M}_i \bar{M}_j - \frac{1}{3} \delta_{ij}) \quad (5)$$

12. Savage, H., Clark, A., and Powers, J., IEEE Trans. Mag. MAG-11, 1355.
13. Klimker, H., Rosen, M., Dariel, M. P., and Atzmony, U., Phys. Rev. B10, 2968 (1974).
14. Blessing, G., Cullen, J., and Rinaldi, S., to be published.

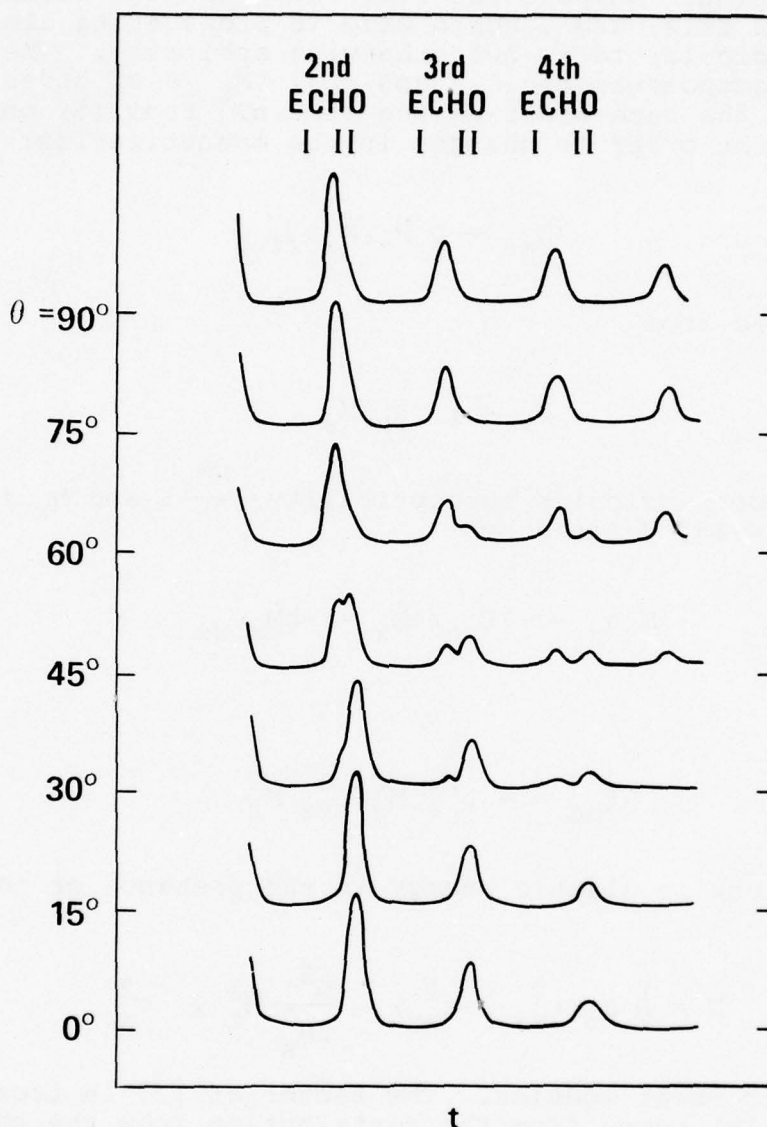


Fig. 6 Observed echo pattern for shear waves propagating perpendicular to the applied field H in amorphous $TbFe_2$. The changes in the amplitudes of the peaks as the angle θ between the direction of H and the direction of polarization is varied indicate the presence of two normal modes with polarization parallel and perpendicular to H . The positions of the peaks do not change, indicating that there is no variation in the velocity of the normal modes.

The \bar{M}_i are the components of the average magnetization normalized to one at saturation. Suppose the field and thus the average moment to be along the z axis, and a sound wave is propagating along x, polarized perpendicular to x, but otherwise arbitrary. The only non-zero strain components are ϵ_{xy} and ϵ_{xz} . \bar{M}_z is of order one; \bar{M}_x and \bar{M}_y are of the same order as the strain. From (5) only ϵ_{xz} is coupled in first order to changes in the magnetization:

$$\bar{E}_{me} \approx b \bar{M}_z \bar{M}_x \epsilon_{xz} \quad (6)$$

\bar{M}_x can be obtained from

$$\bar{M}_x = \chi h_x / M_s \quad (7)$$

where χ is the perpendicular susceptibility $(\frac{dM_x}{dh_x})$ and h_x is the magnetoelastic field, defined by

$$M_s h_x = -\partial \bar{E}_{me} / \partial \bar{M}_x = -b \bar{M}_z \epsilon_{xz} \quad (8)$$

Thus

$$\bar{E}_{me} = -b^2 \chi \bar{M}_z^2 \epsilon_{xz}^2 / M_s^2 \quad (9)$$

The total increment in elastic energy in the presence of the sound wave is then

$$\bar{E} = \frac{1}{2} \mu_0 (\epsilon_{xz}^2 + \epsilon_{xy}^2) - \frac{b^2}{2M_s^2} \bar{M}_z^2 \chi \epsilon_{xz}^2 \quad (10)$$

μ_0 is the elastic shear modulus. The factor of 1/2 in front of the second term in (10) comes from the contribution from the change in magnetic energy, which is $-1/2 \bar{E}_{me}$. From (10) we see that the degeneracy of the shear modes is broken. The normal modes are waves, linearly polarized along the z and y axes, i.e., parallel and perpendicular to the field. These conclusions moreover do not depend for their validity on the simplified model just presented. Because of elastic isotropy in the plane perpendicular to the propagation only the magnetic field picks out a special direction, and we expect the normal modes to be polarized either parallel or perpendicular to the field. This principle applies to any elastically isotropic system in the presence of a magnetic field; the size of the splitting is proportional to the square of the magnetoelastic coupling. Since b is close to $b_2/2$, which, as we mentioned at the outset is two orders of magnitude larger than the coupling in any other room-temperature

magnet, it is not surprising that this effect has never been reported before in polycrystals. The same effect of course can take place in single crystals if the propagation is for example parallel to a [100] direction in a cubic crystal. Again, elastic isotropy of the (100) plane can be broken by a field in this plane. This effect, magnetoacoustic birefringence, was predicted [15] essentially using Equation (3), and has been observed in single crystals of Ni and Fe_3O_4 [16]. In [16], velocity differences of the order of 10^{-3} were measured using a phase-sensitive technique. We have observed relative changes ($\Delta v/v$) of twenty-six percent in $\text{Sm}_{.7}\text{Ho}_{.3}\text{Fe}_2$ (Table 1).

Magnetic field dependences of both velocities were separately measured to 25 kOe. Results are plotted for all four samples in Figure 7. The splitting into two waves was clearly observable to fields of 25 kOe, the largest available.

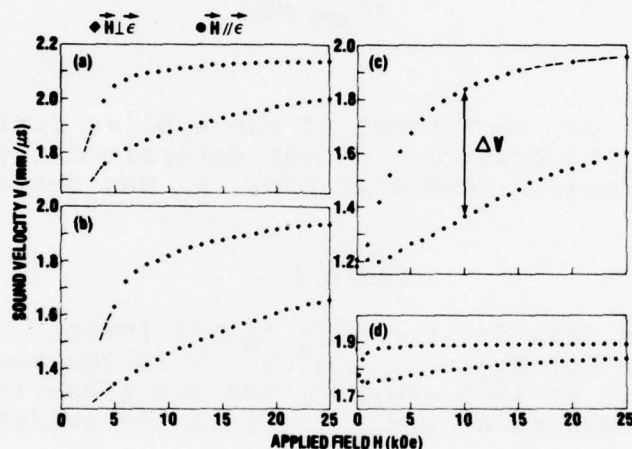


Fig. 7 Velocity as a function of applied field for $\vec{H} \perp \vec{E}$ and $\vec{H} // \vec{E}$ in (a) $\text{Tb}_{.3}\text{Dy}_{.7}\text{Fe}_2$, (b) $\text{Sm}_{.88}\text{Dy}_{.12}\text{Fe}_2$, (c) $\text{Sm}_{.7}\text{Ho}_{.3}\text{Fe}_2$, and (d) $\alpha\text{-TbFe}_2$.

The velocities of the slow modes could not be saturated even at 25 kOe; the velocity of the fast mode of $\alpha\text{-TbFe}_2$ is apparently saturated above 2 kOe (Figure 7d). The fast modes of the polycrystals, especially the compounds containing Sm show significant field dependencies to 25 kOe. By plotting the modulus of the fast modes versus $1/H$ we obtained the pure elastic modulus μ_0 from the extrapolation $1/H \rightarrow 0$ (Figure 8). The values so obtained are listed in

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16. Lüthi, B., Appl. Phys. 8, 107 (1966).

Table 1. Then, from the slopes of $(\mu_0 - \mu)^{-1}$ vs. H plots (Figure 9), we obtained a pair of coupling parameters $b_{//}$ and b for the slow and fast modes respectively for each sample. These are also given in Table 1.

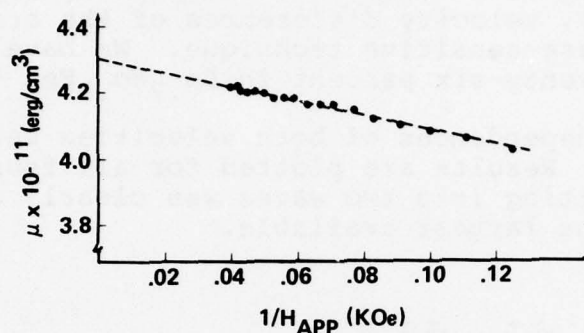


Fig. 8 Modulus (μ) vs. reciprocal of the applied field for $\text{Tb}_{.3}\text{Dy}_{.7}\text{Fe}_2$ polycrystal. μ was determined from the velocity of the fast mode. From this plot, μ_0 was determined.

TABLE 1

Elastic and magnetic data for the RFe_2 alloys investigated. The saturation magnetization M_s is in emu/cm^3 ; V is the sound velocity; elastic modulus μ_0 is in 10^{11} erg/cm^3 ; and the effective magneto-elastic coupling constants b and $b_{//}$ are in 10^9 erg/cm^3 .

RFe_2 Alloy	M_s	$(\frac{\Delta V}{V})_{\max}$	μ_0	b	$b_{//}$	$(\frac{\Delta V}{V})$
$\text{Tb}_{.3}\text{Dy}_{.7}\text{Fe}_2$	780	0.13	4.3	0.13	1.3	0.01
$\text{Sm}_{.88}\text{Dy}_{.12}\text{Fe}_2$	360	0.21	3.4	0.47	1.4	0.11
$\text{Sm}_{.7}\text{Ho}_{.3}\text{Fe}_2$	145	0.26	3.5	0.35	0.9	0.15
a- TbFe_2	390	0.06	2.9	0.07	0.5	0.02

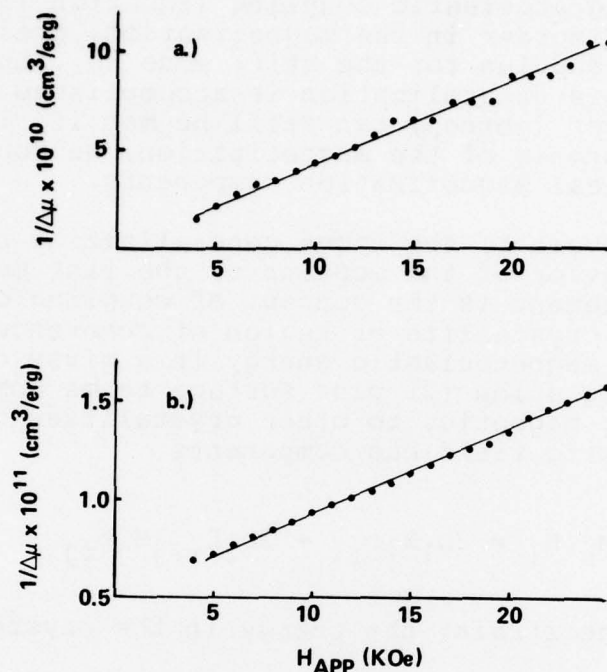


Fig. 9 Reciprocal of the difference of the modulus from the pure elastic modulus ($\mu - \mu_0$) vs. magnetic field for $Tb_{0.3}Dy_{0.7}Fe_2$. From the slopes the b's were determined. a) soft mode, b) stiff mode.

As far as the slow ($\vec{H} \parallel \vec{c}$) modes are concerned, the simple model outlined above describes the data fairly well. Since for large H , $\chi \approx M_s/H$, Equation (10) predicts for the change in modulus $\delta\mu_{//}$ of the slow mode

$$\delta \mu_{//} = - b_{//}^2 / M_s H \quad (11)$$

Equation (11) correctly gives the large H behavior of the slow mode. It also predicts that the modulus changes are larger, for a given b , for samples with smaller saturation magnetization, in agreement with the facts; the largest reduction in modulus occurs in $Sm_{0.7}Ho_{0.3}Fe_2$, which has the lowest M_s (Table 1). (Table 1 also shows that the b 's are roughly the same for all the polycrystals.)

On the other hand, the model is inadequate for a description of the field dependence of the fast mode. A generalization, based again on isotropic magnetoelastic coupling (Equation (5)) but taking some account of the disorder in the magnetization, predicts that there is a change in modulus for the stiff mode $\delta\mu$, but going as $1/H^3$ for large H . This generalization is accomplished by recognizing that the requirement of isotropy can still be met if, instead of averages of the components of the magnetization, we couple the strains to products of the local magnetization components.

Although an improvement, the above generalization cannot reproduce the observed $1/H$ behavior of the modulus of the fast modes. The key to further improvement is the concept of coupling on a local scale, i.e. for each crystallite or region of coherence in the amorphous case. The magnetoelastic energy in a given cubic crystallite is just Equation (2) plus surface terms coming from couplings, elastic or magnetic, to other crystallites or regions. The local magnetoelastic field has components

$$M_s h_i = 2b_1 M_i \epsilon_{ii} + 2b_2 \sum_{i \neq j} M_j \epsilon_{ij} \quad (12)$$

To second order in the strain, the energy in the crystallite is lowered by

$$\delta E = -\frac{1}{2} \sum_{i,j} h_i \chi_{ij} h_j \quad (13)$$

The h_i are components of a real vector only when $b_1=b_2$; in general their value depends on the set of axes defining them. Thus even if the magnetization and the strain were uniform the h_i would vary from crystallite to crystallite. The material has its total energy lowered by the interaction (Equation (13)) at the expense of inducing a disorder in the magnetization. Therefore at very high magnetic fields, in the presence of a strain there are fluctuating components of the magnetization in the plane perpendicular to the net magnetization direction; these in turn will couple to the strain components in this plane. This is the mechanism which leads to a $1/H$ dependence of the modulus of the stiff mode.

The arguments given above should hold true even if there are interactions between the grains or local regions, just as long as these do not completely inhibit the creation of magnetization fluctuations via the magnetoelastic interaction. For the limiting case of independent crystallites it is possible to calculate the change in modulus of either mode at high fields. Assuming a uniform distribution of crystallite axes, δE (Equation (13)) is averaged

over the unit sphere for a set of fixed strains. The results for the changes in shear moduli are

$$\delta\mu_{\perp} \approx -\frac{8}{105} \frac{(b_2-b_1)^2}{HM_s} \equiv -\frac{b^2}{HM_s} \quad (14)$$

for polarization perpendicular to the field (stiff mode) and

$$\delta\mu_{//} \approx -(1+\frac{4}{5} \frac{b_1-b_2}{b_1+b_2} + \frac{4}{15} (\frac{b_1-b_2}{b_1+b_2})^2) \frac{(b_1+b_2)^2}{HM_s} \equiv -\frac{b_{//}^2}{HM_s} \quad (15)$$

for the polarization parallel to the field (soft mode). As we anticipated, the $1/H$ behavior of the fast mode depends entirely on the anisotropy in the single crystal magnetostriction. In fact, the approach to saturation magnetostriction was previously shown [17] to behave in a similar way. As we mentioned in the introduction, it is characteristic of RFe_2 materials that $\lambda_{111} \gg \lambda_{100}$ ($b_2 \gg b_1$).

In the large anisotropy limit, i.e. $b_1 \rightarrow 0$ from (14) and (15) the ratio of the coefficients of the $1/H$ law for the two modes is predicted to be

$$\left(\frac{b_{\perp}}{b_{//}}\right)^2_{b_2 \rightarrow 0} = 0.163 \quad (16)$$

In the limit of magnetoelastic isotropy, i.e. $b_1=b_2$, the prediction is that $b_{\perp}/b_{//} = 0$. The experimental values listed in Table 1 all fall between these two extremes of the theory. From the discussion following Equation (13) we take this to indicate the presence of interactions between grains. However this intergranular interaction is not strong enough to eliminate the magnetoelastic anisotropy. Metallographic work shows that the samples containing Sm , which according to Table 1 are closest to behaving like collections of independent crystallites, contain appreciable amounts of intergranular precipitates. Much finer grain boundaries were observed in the $Tb_{.3}Dy_{.7}Fe_2$ sample.

There are aspects of this work on disordered RFe_2 materials still to be worked out, including a description of the low field behavior of the moduli and measurements of their temperature dependencies. The most important result of this section and which leads us into the next, is the breaking of the elastic isotropy by the magnetoelastic coupling.

17. Lee, E. W., Proc. Phys. Soc. A 67, 381 (1954).

RESONANT TRANSMISSION OF SOUND IN A SINGLE CRYSTAL

Up to this point there has been a certain dichotomy in the presentation; on the one hand we described single crystal results in terms of changes in velocity of modes (C_{44} , C_{11} - C_{12}) of fixed polarization while on the other we interpreted polycrystalline data by means of modes of constant velocity whose polarization depended on the direction of the field. In describing the single-crystal results we have implicitly assumed that the appropriate symmetry

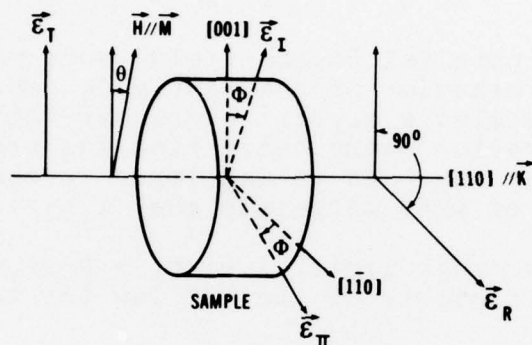


Fig. 10 Experimental arrangement for the resonance experiment in $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_2$ single crystal. A shear wave is generated by a transmitting transducer with polarization $\vec{\epsilon}_T // [001]$ and propagates through the sample along the $[110]$ direction. At the other side of the sample a receiving transducer with polarization $\vec{\epsilon}_R // [1\bar{1}0] \perp \vec{\epsilon}_T$ detects a signal due to the rotation ϕ of the polarizations of the normal modes ($\vec{\epsilon}_I$, $\vec{\epsilon}_{II}$) induced by magnetoelastic interaction with the magnetization \vec{M} . The magnetization is assumed parallel to the field \vec{H} applied in the (110) plane at an angle θ from the $[001]$.

for elastic modes was determined by the crystal. Clearly there is some competition between the crystalline symmetry and that induced by the magnetic field via the magnetoelastic coupling. Generally the latter is taken to be a perturbation of the former [3,8], so that the directions of polarization of the transverse modes are only slightly tilted by an angle ϕ from the crystalline axes for any orientation θ of the magnetic field with respect to the $[001]$ axis. (See Figure 10). This tilting of the polarization is predicted to occur only if both b_2 and b_1 (Equation (1)) are non-zero. In fact a straightforward but lengthy calculation of the velocities and

directions of polarization of the normal modes can be carried out; [8, 18] the resulting equations for the velocities v_{\pm} are:

$$v_{\pm}^2 = \frac{C+C'}{2} \pm \left(\left(\frac{C-C'}{2} \right)^2 + B^2 \right)^{\frac{1}{2}} \quad (17)$$

The parameters C , C' and B are:

$$C \equiv C_{44}^0 / \rho - b_2^2 \cos^2 \theta / (H+H^0) M_S \rho \quad (18)$$

$$C' \equiv (C_{11}-C_{12})^0 / 2\rho - b_1^2 \sin^2 \theta / (H+H^0) M_S \rho \quad (19)$$

$$B \equiv b_1 b_2 \sin \theta \cos \theta / (H+H^0) M_S \rho \quad (20)$$

where ρ is the density.

If $B=0$ ($\theta=0$ or 90°) there are two transverse waves polarized along $[001]$ and $[1\bar{1}0]$ with velocities $C^{1/2}$ and $C'^{1/2}$ respectively; though either C or C' is reduced (depending on whether $\theta = 0$ or 90°) by the magnetoelastic coupling, they are nonetheless "pure" modes in that they are polarized along crystalline axes. In general we only expect strong effects if $C \approx C'$ assuming b_1 and $b_2 \neq 0$. We know from just looking at the curves of Figure (2) that this will happen for some $\theta = \theta_c$ in $Tb_{.3}Dy_{.7}Fe_2$ because $C < C'$ for $\theta=0$ but $C > C'$ for $\theta=90^\circ$. (θ is measured with respect to $[001]$). Furthermore, $b_1 \ll b_2$ in RFe_2 compounds so the range of θ around θ_c where strong coupling can take place is small. We have plotted v versus θ in Figure (11) using known values of b_2 , M_S , C_{44} , $C_{11}-C_{12}$ and H_0 and taking $b_1 \approx 0.1 b_2$, $H=8$ kOe. To see what happens to the polarization direction under these conditions, we calculated ϕ from

$$\tan \phi_{\pm} = \frac{B}{(C'-C)/2 \pm \left((C'-C)^2/4 + B^2 \right)^{\frac{1}{2}}} \quad (21)$$

ϕ is very close to 0 or 90° except in the immediate vicinity of θ_c where it changes abruptly.

To check this "pulling" of the axes of polarization off the crystalline axes we performed a transmission experiment (Figure (10)) in which receiving and transmitting transducers were arranged with linear polarization along crystallographic axis and orthogonal to one another. A magnetic field of 8 kOe was rotated in the (110) plane, and the output of the receiver recorded. We expect this output to vary as $A \sin 2\phi$, where A is another oscillatory function of θ coming from interference. Using the same set of parameters listed above, we have calculated the expected amplitude as a function

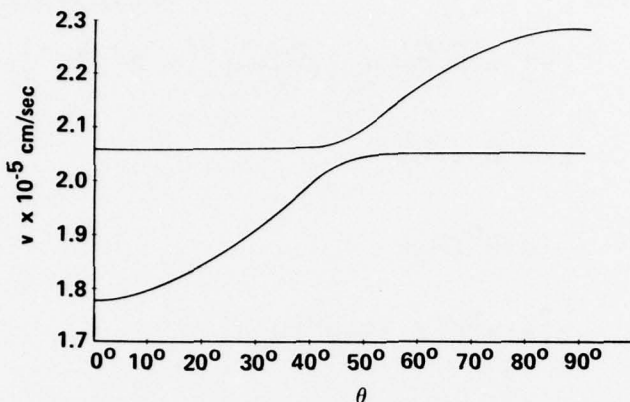


Fig. 11 Dispersion in the velocity of the shear normal modes for [110] propagation in $\text{Tb}_3\text{Dy}_7\text{Fe}_2$ caused by the interaction of the strain with the magnetic moment in the (110) plane at an angle θ from the [001]. The parameters used are: $b_1 = 2 \times 10^8 \text{ erg/cm}^3$; $b_2 = 2.3 \times 10^9 \text{ erg/cm}^3$; $(C_{11}-C_{12})/2 = 3.9 \times 10^{11} \text{ erg/cm}^3$; $C_{44} = 4.8 \times 10^{11} \text{ erg/cm}^3$; $M_s = 1000 \text{ emu/cm}^3$; $\rho = 9.2 \text{ g/cm}^3$, $H_0 = 20 \text{ kOe}$, $H = 8 \text{ kOe}$.

of θ . The result is plotted in Figure (12a). The envelope, peaked around 45° , is due to $\sin 2\phi$ while the oscillations come from A . We have also plotted a modified version of the amplitude obtained by correcting Figure (12a) for depolarization and finite resolution: Figure (12b). The experimental results are given in Figure (12c). There is reasonably good agreement, i.e. the theory reproduces the sharp central peak and much of the structure away from the crossover angle. Although this agreement verifies our picture of the rotation of the polarization, the theoretical model must still be made consistent with our earlier discussion of Figure (2) in which we pointed out that the slight stiffening of the $C_{11}-C_{12}$ mode was inconsistent with Equation (2). Including non linear terms of the form of Equation (4) means simply replacing $b_1^2/M_s(H+H_0)$ by itself plus f_1 and $b_1b_2/M_s(H+H_0)$ by itself plus f_2 in Equation (19) and (20) [18]. f_1 must be negative and larger in magnitude than $b_1^2/M_s(H+H_0)$ to explain the stiffening of $C_{11}-C_{12}$. Since the relative change in $C_{11}-C_{12}$ is about 2%, f_1 must be about 10^{10} erg/cm^3 . Now the width of the central peak in the transmission amplitude is best fit by choosing $b_1b_2/M_s(H+H_0) + f_2 \approx 10^{11} \text{ erg/cm}^3$. If $|f_1| \approx |f_2|$ it must be the combination b_1b_2 that is responsible for coupling the

18. Rinaldi, S., and Cullen, J., to be published.

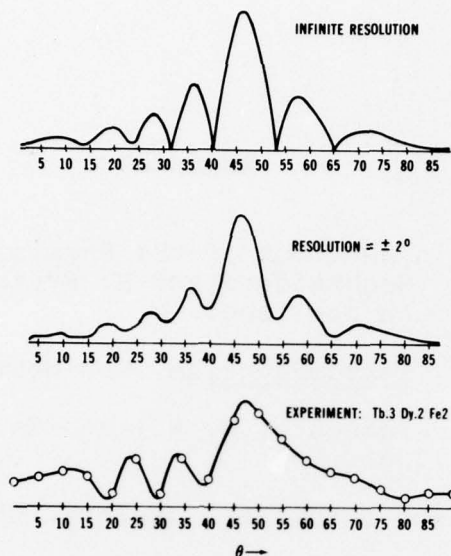


Fig. 12 Comparison between the theoretical prediction and the experimental results of the resonance experiment described in the text and in Figure 10. The parameters used for the theoretical curves are the same as in Figure 11.

two transverse modes. At this stage of our knowledge, we can only regard these estimates as tentative.

The expectation of large changes in elastic properties of RFe_2 compounds due to moment rotations has been amply justified. The measurements described here have further shown that the elastic isotropy of polycrystalline and amorphous RFe_2 alloys is removed in a magnetic field and that the elastic anisotropy of crystalline compounds can be nullified and even inverted. Most of the observed effects are accounted for by linear magnetoelastic theory if $b_2 \gg b_1$ and proper account of the fluctuation in magnetization is taken. Discrepancies indicate the presence of non linear magnetoelastic interactions.

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